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Half-Life of the Long-Lived Tc^{99}

SHERMAN FRIED, A. H. JAFFEY, N. F. HALL,* AND L. E. GLENDENIN
Chemistry Division, Argonne National Laboratory,† Chicago, Illinois

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The half-life of Tc^{99} has been determined on materials isolated both from fission products and from irradiated molybdenum. The determinations were made on aliquots of solutions prepared by dissolving weighed portions of compounds of known composition.

The counts were made absolute by comparing them with Co^{60} and RaDEF standards. These determinations resulted in a value of 2.12×10^{5} years for the half-life of Tc^{99}.

A detailed description of the absolute beta-counting techniques is given.

I. INTRODUCTION

TECHNETIUM, element number 43, long a missing element in the periodic system, was discovered through its radioactivity by Perrier and Segrè in 1937 as a product of the bombardment of molybdenum by deuterons.[1]

Various other investigators[2, 3] demonstrated the occurrence of this element in fission products as the radioactive isotope Tc^{99}. This isotope can also be prepared[4, 5] by irradiation of natural molybdenum in a chain reacting pile, where it is formed by the reaction

$$Mo^{98}(n,\gamma)Mo^{99}\xrightarrow[67\text{ hr}]{\beta}Tc^{99}. \qquad (1)$$

Other isotopes of technetium have been prepared by various methods[6] and now the mass range from Tc^{92} to Tc^{105} is known with more or less certainty.[7]

The isotope Tc^{99} is the most suitable one for chemical investigation because of its long half-life, variously estimated as 2×10^{5} to 9×10^{5} years. These values were based either on yield estimates or on an assumed formula for a particular compound. Since early workers in the field had little information as to the chemical formulas of technetium compounds, these measurements were correspondingly uncertain.

Recently, some aspects of the chemistry of technetium have been elucidated and several definite compounds have been prepared and identified;[8] and it was with this new information in mind that it was proposed to re-examine the half-life determination of Tc^{99} and, if possible, to reconcile the various values given in the literature. It was decided to make the determinations on technetium isolated from pile-irradiated molybdenum as well as from material from fission products. Because of the high fission yield of Tc^{99} and the sufficiently high neutron capture cross section of Mo^{98}, enough material was available from both sources to make reasonably accurate weighings of compounds possible.

The absolute counting necessary for a specific activity determination was done relative to coincidence-counter standardized Co^{60} samples. The Co^{60} beta-radiation has almost the same energy and aluminum absorption curve as Tc^{99}, which simplifies the problem of geometry standardization.

II. PURIFICATION OF TECHNETIUM AND PREPARATION OF TECHNETIUM SAMPLES

The details of the purification of technetium from gross amounts of impurities and from radiochemical contaminants are to be described in a paper devoted to

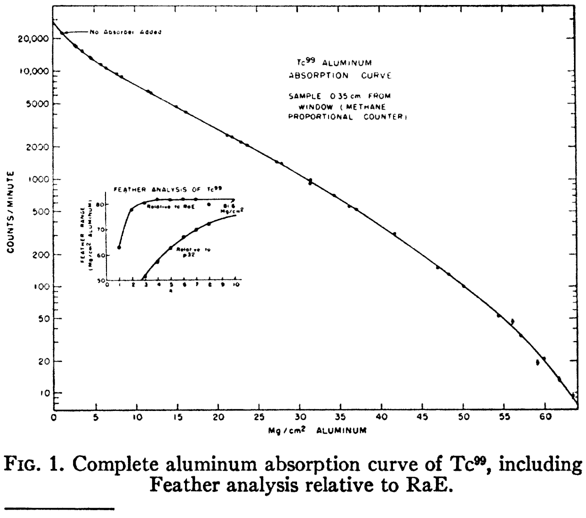

FIG. 1. Complete aluminum absorption curve of Tc^{99}, including a Feather analysis relative to RaE.

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* On leave of absence from the University of Wisconsin, Madison, Wisconsin.

† The material presented here was given in part in a paper by Sherman Fried and Norris F. Hall presented at a meeting of the American Chemical Society, April, 1950.

[1] C. Perrier and E. Segrè, J. Chem. Phys. **5**, 712 (1937).

[2] D. C. Lincoln and W. H. Sullivan, "Discovery of the $10^{6}y$. Tc^{99} isotope. I." Paper 7.17.2, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Co., Inc., New York, to be published), National Nuclear Energy Series, Division IV, Vol. 9B; Hanford Report No. (H)CN-3449, October 25, 1945, unpublished.

[3] R. P. Schuman, "Discovery of the $10^{6}y$. Tc^{99} isotope. II.," Paper 7.17.3, *Radiochemical Studies: The Fission Products;* Metallurgical Laboratory Report No. CC-3434, February 16, 1946, unpublished.

[4] E. E. Motta and G. E. Boyd, Oak Ridge National Laboratory Report MonC-169, September 15, 1946, unpublished.

[5] Motta, Boyd, and Larson, Phys. Rev. **72**, 1270 (1947).

[6] G. T. Seaborg and I. Perlman, Revs. Modern Phys. **20**, 585 (1948).

[7] Technetium isotopes of masses 103 and 104 have not yet been found.

[8] Paper presented at a meeting of the American Chemical Society, April, 1950, by S. Fried and N. F. Hall.

the chemistry of the element; but, in brief, the purification involved the reduction of technetium sulfide to the metal by action of hydrogen[9] at 1000–1100°C. Subsequent high temperature oxidation of the metal to the oxide by oxygen allowed it to be sublimed away from impurities, since the oxide is quite volatile.

The aluminum absorption curve shown in Fig. 1, which agrees with those found by other workers in the field,[2–5] shows the absence of radioactive contaminants. Spectrographic analysis of solutions of technetium prepared by this method indicated no gross impurities.[10]

The compound NH_4TcO_4 has been prepared[8] by action of a mixture of ammonium hydroxide and hydrogen peroxide on technetium oxide. The formula of the compound was deduced by Prof. W. H. Zachariasen using x-ray diffraction analysis.[11] It is soluble in water and is nonhygroscopic. Since it contains a known proportion of technetium, it can serve as a gravimetric standard for the preparation of solutions of known concentration of technetium for use in beta-counting.

Technetium metal itself also serves as a gravimetric standard, but more elaborate methods must be employed. The metal cannot be directly dissolved in a suitable solvent.[12] To dissolve the technetium, a weighed sample of the metal was converted quantitatively to the volatile oxide (presumably TcO_3), and was then washed out of the reaction tube into a calibrated volumetric flask. In this way the concentration of technetium in solution was determined directly from the initial weight of the metal. The equations representing the sequence of reactions probably are:

$$\mathrm{Tc}\xrightarrow[400\text{–}600^\circ\mathrm{C}]{O_2}\mathrm{Tc\ oxide\ }(TcO_3)\xrightarrow{NH_4OH+H_2O_2} NH_4TcO_4\ \text{(in solution).} \qquad (2)$$

Aliquots were withdrawn from the standardized solutions by means of calibrated micro-pipettes.[13] To minimize beta-particle back-scattering, quantities of the solutions of 9.97 and 25.02 microliters were placed on films of Tygon ($\sim$0.05 mg/cm^2) or rubber hydrochloride[14] ($\sim$0.6 mg/cm^2) supported by aluminum rings. The measured droplets were allowed to dry in air at room temperature. The weight of technetium deposited in this manner was 0.5 to 1.5 μg, spread over an area of about 0.2 cm^2. Precision counting of the samples showed that the aliquoting technique gave results reproducible to about 0.3 percent. From the weight of technetium in a sample and its counting rate, a relative specific activity could be determined, requiring only the determination of the counter geometry to make it an absolute value.

Sulfur in Technetium

Although spectrographic analyses showed no large amount of impurities, the first result for the relative specific activity of the molybdenum-derived technetium was 25 percent lower than that of the fission product material. A sulfate test with barium chloride on the former solution showed that this discrepancy was due to the presence of sulfur in the technetium metal. The presence of any appreciable quantity of sulfur of course introduces a gravimetric error, which tends to decrease the apparent specific activity and hence increase the measured value of the half-life. Because of the difficulty of detection, this impurity was missed in the spectrographic analyses. The preparation of the metal in a fairly pure state was found to require prolonged action of hydrogen at high temperatures.

Fortunately an independent method for the determination of technetium was developed. The absorption spectrum of Tc(VII) in perchloric acid solution showed characteristic intense bands in the ultraviolet region[15] at 2440 and 2490A. Since the absorption follows Beer's Law, it was possible to determine the concentration of technetium after having once determined the molar extinction coefficient of a known standard solution.

The molybdenum-derived technetium was made into metal again, and the specific activity redetermined. The result was still 4 percent lower than that of the fission-product technetium; but absorption spectrum analysis, standardized with fission-product technetium, showed the concentration of the solution to be 94.5 ($\pm$1.0) percent of that calculated from the weight of the metal. The relative specific activity of the molybdenum-derived technetium thus agreed within experimental error with that of the fission-product material.

Although this discrepancy was removed, it must be noted that the presence of small amounts of sulfur in the fission-product technetium cannot be entirely ruled out, although a text with barium chloride gave no precipitate. However, another aspect of the present measurements makes this possibility unlikely. Following purification, the fission-product material was twice passed through the cycle: Tc sulfide→Tc metal→Tc oxide→NH_4TcO_4 solution→specific activity determination. In the first run, the solution concentration was determined by weighing the metal; in the second run, the solution was evaporated and the NH_4TcO_4 was weighed. The two specific activities determined from these independent weighings checked closely (Table II). Since it is very unlikely that the amounts of sulfur

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[9] S. Fried, J. Am. Chem. Soc. **70**, 442 (1948).

[10] These analyses were carried out by Mr. John P. Faris of the Analytical Group of this Laboratory.

[11] The use of x-ray diffraction as a method of determining formulas of technetium compounds has proved extremely fruitful in the elucidation of the chemistry of the element.

[12] The metal dissolves in 8*M* HNO_3, but this is not considered suitable for the preparation of counting samples. It was feared, at the present state of knowledge of Tc chemistry, that loss of Tc would result from drying acid solutions. All counting samples were made from neutral or alkaline solutions of Tc salts.

[13] The technique is described in P. L. Kirk, *Quantitative Ultramicro Analysis* (John Wiley & Sons, Inc., New York, 1950).

[14] Reed Laboratories, Akron, Ohio.

[15] The absorption spectrum of Tc(VII) and analyses of the Tc solutions were determined by J. C. Hindman and Philip Wehner of this Laboratory with a Cary Recording Spectrophotometer.

carried through these runs were in just the right proportions to give apparent agreement, we conclude that the fission-product material was essentially free of sulfur impurity.

III. PREPARATION AND CALIBRATION OF STANDARD SAMPLES

The Co^{60} standards, each containing less than a microgram of cobalt, were prepared from Oak Ridge material having a specific activity of 300 mc/g. To minimize back-scattering, the material was deposited on rubber hydrochloride films identical with those used for the technetium samples. The beta-activities of the first samples prepared were found to have decreased after several months, apparently due to the deposition of water on the hygroscopic cobalt nitrate. The formation of the insoluble CoS by evaporation of the deposited droplet in an atmosphere of H_2S served to eliminate the difficulty. However, since this salt tended to concentrate in a narrow ring, the cobalt was kept spread during evaporation by the addition of a small amount of glue. The final samples were spread over an area of about 0.3 cm^2, and contained approximately 4×10^5 disintegrations per minute (dis/min).

The absolute disintegration rates of these samples were determined by coincidence counting of β's and γ's in the usual manner.[16] The simplicity of the Co^{60} disintegration made possible a calibration free of many of the difficulties occurring with complex decay schemes. Co^{60} disintegrates[6] through the emission of a 310-kev beta-particle, followed by two gamma-rays in cascade (1.17 and 1.33 Mev). The disintegration rate N was calculated from the expression

$$N = N_\beta N_\gamma / C_{\beta\gamma}, \tag{3}$$

where N_β = singles counting rate due to the Co^{60} beta-particles in one detector, N_γ = singles counting rate due to Co^{60} gamma-rays in the other detector, and $C_{\beta\gamma}$ = coincidence rate. N_β was corrected for background, for gamma-ray counting rate in the beta-particle detector, and for resolution losses; N_γ was corrected for background and resolution losses; $C_{\beta\gamma}$ was corrected for accidentals, gamma-gamma-coincidences, cosmic-ray background, and resolution losses in both channels. All corrections were relatively small. Since the conversion coefficients of the Co^{60} gamma-rays have been found to be very small,[17] no correction was necessary for conversion electrons.

The flow-type atmospheric proportional counters (methane or argon-10 percent methane) used as detectors were shaped like the usual bell-shaped mica-window G-M tube, except that a loop of 1-mil wire served as center electrode. The beta-counting tube had a 0.6 mg/cm^2 window of rubber hydrochloride. A 300 mg/cm^2 aluminum absorber was used over the gamma-counting tube to cut out beta-particles and to act as a radiator for increasing the gamma-ray efficiency. Highly active sources gave sufficiently high coincidence rates without the use of more efficient gamma-counters. It was possible to use such active sources only because of the high singles counting rates attainable with the beta-counting proportional counter.

When a methane containing tube was used at about 1 mv sensitivity, the plateau for Co^{60} betas started at 2600 volts; with the argon-methane mixture, the plateau started at 1750 volts. These voltages were higher than would be the case with other tube designs and were largely necessitated by the small pulses from tracks in the corners of the tube (e.g., near the edge of the window). Since the plateau voltages were relatively high, the fact that the effective dead-time t of the tube increased with voltage for both gases, led to fairly high values for t, about 5 μsec for methane, and 8–12 μsec for argon-methane.

The effective dead-times were measured empirically[18] by the paired-sample technique and corresponded to losses of 0.8 percent at 100,000 counts/min in the methane counter. Counting rates in the argon-methane tube were relatively low, resulting in small corrections.

The proportional counters, mounted coaxially, were placed with windows as close together as possible, leaving only enough space between them for absorbers and sample. For determining the gamma-gamma-coincidence correction and the gamma-ray correction for the beta-ray singles count, an aluminum absorption curve was taken with the beta-ray tube. Because of the large variation of gamma-counting efficiency with absorber thickness, the gamma-ray absorption curve was extrapolated from beyond the beta-range to zero absorber. To minimize the number of apparent gamma-gamma-coincidences due to back-scattered Compton gamma-rays, the sample was sandwiched between two lead absorbers having holes only large enough to ensure both tube windows "seeing" the entire sample.

The signal from each proportional counter passed through a cathode-follower, and then through a cable to the main amplifier. The amplifier input contained a compressor circuit (crystal diode in series with 10,000 ohms) and a differentiating network. Each coincidence channel contained a pulse-height discriminating univibrator (cathode-coupled, first tube normally conducting), operating at a fixed $\frac{1}{2}$-volt sensitivity, which was followed by a similar univibrator with variable pulse width. A modified Rossi circuit was used for mixing. To ensure that all pulses entering the mixer were counted as single counts as well, the pulses to the singles scalers were taken from the second univibrators.

The pulses from the proportional counters were found

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[16] J. V. Dunworth, Rev. Sci. Instr. **11**, 167 (1940).

[17] M. Deutsch and K. Siegbahn, Phys. Rev. **77**, 680 (1950) (for each gamma-ray, $a = 0.0002$).

[18] T. P. Kohman, "General method for determining coincidence corrections of counting instruments," paper 22.50, *The Transuranium Elements* (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Division IV, Vol. 14B.

TABLE I. Co^{60} standards calibrated by coincidence counting (in units of 10^5 dis/min).

	Sample No.	dis/min as of Jan. 1, 1950	Ratio to Co-17 from gamma-count	Co-17 dis/min Jan. 1, 1950	Deviation from mean
Coincidence circuit I	Co-17	$3.83_9\pm0.02_0$	1.000	$3.83_9\pm0.02_0$	0.02_4
	Co-T-II	$2.88_0\pm0.04_0$	$0.735_3\pm0.005_0$	$3.91_7\pm0.06_1$	0.05_4
	Co-T-III	$2.93_1\pm0.05_5$	$0.740_0\pm0.005_3$	$3.96_1\pm0.08_0$	0.09_8
	Co-IV	$4.77_7\pm0.05_9$	$1.221_0\pm0.005_7$	$3.91_2\pm0.04_9$	0.04_9
	Co-VI	$4.85_1\pm0.05_1$	$1.222_5\pm0.005_0$	$3.96_8\pm0.04_5$	0.10_5
	Co-18	$3.79_8\pm0.03_4$	$0.998_2\pm0.004_3$	$3.80_5\pm0.03_8$	0.05_8
	Co-19	$3.85_0\pm0.01_9$	$0.998_6\pm0.004_7$	$3.86_3\pm0.02_6$	0.00_0
			Weighted mean	$3.86_3\pm0.01_3$	
Coincidence circuit II	Co-17	$3.83_4\pm0.04_0$	1.000	$3.83_4\pm0.04_0$	0.01_3
	Co-19	$3.81_8\pm0.01_9$	$0.998_6\pm0.004_7$	$3.82_3\pm0.02_6$	0.02_4
	Co-VI	$4.73_4\pm0.02_1$	$1.222_5\pm0.005_0$	$3.87_2\pm0.02_3$	0.02_5
			Weighted mean	$3.84_7\pm0.01_6$	

to have a relatively large range of lagging-times, probably due to a mixture of two effects: (1) detector lag-time due to variations in transit-time of the original, ionization-formed electrons through the detector gas (atmospheric pressure), and (2) variation in tripping time of the pulse-height discriminating univibrator due to wide range of pulse heights. A plateau of coincidence rate *vs* resolving time (τ) was determined, and measurements were normally taken well on the plateau, at $\tau=1.3$ μsec. Despite the large value for τ, corrections for accidental coincidences were relatively small.

A number of samples were measured at various times. Table I includes the data on seven samples, corrected for decay to a standard time using the half-life value[6] of 5.3 years. All of the samples were compared in relative activity by gamma-counting to Sample-Co-17, and this sample was used as the primary standard.

Because of the disagreement (see below) between the geometry determined by these Co^{60} standards and that measured with a RaE standard, a number of measurements were attempted with other coincidence systems. Another type of coincidence circuit was used[19] in conjunction with detectors consisting of anthracene crystals and liquid nitrogen cooled photo-multipliers (1P21).

The disintegration rate, N [Eq. (3)] was measured as a function of the photo-multiplier voltages (using a fixed pulse-height discriminator). If the only effect of changing these voltages were to change the counter efficiencies (essentially by varying the pulse-height selection level), the value of N calculated from Eq. (3) would be expected to be independent of the photo-multiplier current gain. It was found, however, that N was a function of the multiplier voltages. Between 1250 and 1400 volts, a voltage plateau existed giving values of N about 20 percent higher than those in Table I. As the voltage dropped, N decreased until another sloping plateau was found between 750 and 800 volts giving N values checking with those in Table I. Since it is difficult to see why $C_{\beta\gamma}$ [Eq. (3)] should be too low at high photo-multiplier voltages, or too high at low voltages, it seems reasonable to assume that the high N values were due to spurious counts in the singles channels. Other evidence in this laboratory and elsewhere indicates that such spurious counts do occur.

The calibration was repeated some months later with the same proportional counters and amplifiers but with another coincidence circuit,[20] which used Schmitt trigger pairs for pulse-width setting and for mixing. The results shown in Table I may be seen to check very closely with the previous values.

Since there were available in the laboratory some beta-counting standards[21] containing RaDEF mounted on thin rubber hydrochloride films, an attempt was made to check them against the Co^{60} standards. However, because of the difference in energies, extrapolation to zero absorber was quite different in the two samples, making it impossible to correct for the relatively large absorption (air and window = 1.1 mg/cm^2) of the methane proportional counter. The samples were compared in a low absorption counter[22] having a gas and window absorption of 0.1 mg/cm^2. At the pressure used (2 cm of argon—10 percent propane), the RaDEF standard consistently gave geometry values about 18 percent lower than those derived from the Co^{60} standards.

When checking the coincidence measurements showed no error in the Co^{60} standardization, it was suspected that at the low pressure used there might actually be differences in the effective geometries of beta-emitters, depending upon the beta-energies. Such differences might arise from the variation in specific ionization with electron velocity and the greater probability that a fast electron might not ionize in the corner of the tube bordered by window and cylindrical wall.

To check this possibility, another type of low absorption counter[23] was used. It contained a proportional counter similar to the methane counter described above. A gas of relatively high density (argon—10 percent methane) was used at atmospheric pressure to increase the probability of ionization, so that electrons of all energies would be counted. The window was a multilayer film of Tygon with surface density ranging from 0.05 to 0.15 mg/cm^2. To decrease the gas absorption between sample and window, helium was allowed to flow through the containing Lucite box, displacing the air. Samples and absorbers were changed through an opening in the Lucite box; helium flushing followed each such change. Since the sample to window distance was only 3 mm, gas absorption was cut down to 0.05 mg/cm^2. Flat voltage plateaus were measured for both Co^{60} and RaE samples and all counting was done at the

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[19] This measurement was performed by D. W. Engelkemeir of this Laboratory.

[20] Kindly loaned us by D. W. Engelkemeir.

[21] T. B. Novey, Rev. Sci. Instr. 21, 280 (1950).

[22] F. T. Hagemann, "New low absorption GM counter" (report to be issued).

[23] A. H. Jaffey, "Flow-type low absorption proportional counter" (to be published).

same voltage, which lay on both the RaE and Co^{60} plateaus.

Absorption curves were measured for both the RaE and Co^{60} samples and these are shown in Fig. 2. Due to the presence of the RaD beta-particles and RaF (Po^{210}) alpha-particles, it was not possible to measure the extrapolated counting rate of Novey's beta-standards directly. A sample of RaE, separated[24] from RaD and RaF was measured and normalized by comparison with the RaDEF beta-standard at absorber thicknesses exceeding the alpha-particle range. Both absorption curves may be seen to have relatively steep slopes at low absorber thicknesses.

When the small correction for gas and window thickness was made by extrapolating along the aluminum absorption curves, the geometry value using the Co^{60} standard was found to be 42.8 percent, while that from the RaDEF standard was 42.3 percent. The close agreement is fortuitous, since the one percent difference is less than the experimental errors involved in such extrapolations.

The geometry calculated from the dimensions (i.e., from the solid angle subtended by the tube window) agreed very closely with the measured geometry values, indicating that the sensitive volume began immediately in back of the window.

As an additional check on the Co^{60} standardization, the Co^{60} samples were compared with the ampoules of Co^{60} activity issued by the Bureau of Standards and standardized three years earlier. Similar ampoules of a Co^{60} solution were prepared and compared with the Bureau of Standards samples by gamma-ray count, and known aliquots of the solution were mounted on rubber hydrochloride films and compared by gamma-count with our standard Co^{60} samples. Our standardization checked that of the Bureau of Standards within 0.5 percent, which was less than the combined errors of the comparison and the Bureau of Standards standardization.

The close agreement between the various methods of calibrating the Co^{60} standard suggests that its disintegration rate was known to better than 1 percent.

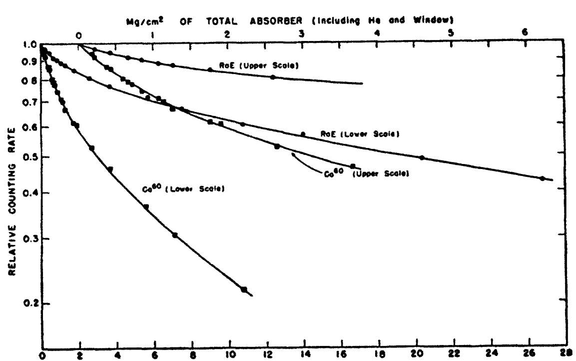

FIG. 2. Absorption curves of RaE and Co^{60} standards in flow type low absorption counter.

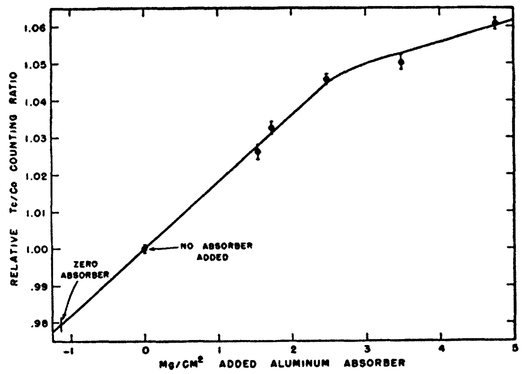

FIG. 3. Ratio of Tc and Co beta-counting rates relative to ratio with no added absorber. Measured with methane proportional counter. Extrapolation to zero absorber corrects for the effect of window and air absorption (1.13 mg/cm²), giving 0.980 for the correction factor.

IV. COMPARISON OF Tc^{99} AND CO^{60} SAMPLES: Tc^{99} HALF-LIFE

In absolute beta-counting by comparison with a standard sample, one of the most likely sources of error is the correction for air and window absorption. This is especially true when the radiation is relatively soft, since the correction is then fairly large. For many radioactivities it is not valid to assume that the aluminum absorption curve extrapolates as a straight line at very low absorber thicknesses; many activities show an upturning of the curve in this region. For example, the Co^{60} and RaE curves of Fig. 2 show this phenomenon. In addition, it is questionable, for large corrections, whether it is valid to assume that the window and air correspond to an equal weight of aluminum. For best results, the comparison between standard and unknown should be done in a low absorption counter, i.e., one in which the gas and window absorption is of the order of 0.1 mg/cm².

In the measurement described here, it was fortunate that the absorption curves for Co^{60} and Tc^{99} were almost superposable, so that an extrapolation of the activity ratios gave rise to only a small correction for air and window. This extrapolation was carried out with the methane proportional counter used for coincidence counting (air and window = 1.13 mg/cm²) and with low absorption counters.[22, 23]

Because the Tc^{99} and Co^{60} absorption curves were so similar, essentially no extrapolation was necessary for the comparisons made with the low absorption counter. The correction was significant, however, for the methane proportional counter. Despite this disadvantage, the methane counter comparisons were more useful because of the greater stability of the instrument as evidenced by the reproducibility of counts. An extrapolation for the methane proportional counter is shown in Fig. 3. The resulting correction factor gives the Tc/Co counting

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[24] This purification was kindly performed for us by T. B. Novey.

TABLE II. Specific activity results by comparison of Tc^{99} and Co^{60} standards.

Counter used	Tc sample No.	Source of Tc[a]	Tc weighed as	Weight of Tc (μg)	Tc/Co ratio of counts/min		Disintegration rate (dis/min) of Co^{60} std. $\times 10^{-5}$	Disintegration rate (dis/min) of $Tc^{99} \times 10^{-4}$	Tc^{99} specific activity dis/min-μg $\times 10^{-4}$
					No absorber added	At zero absorber			
Methane proportional counter	1	F.P.	NH_4TcO_4	$1.54 \pm 0.01_5$	0.1593±0.0002	0.1561±0.0005	3.69 ±0.01	5.76 ±0.03	3.74±0.04
	2	F.P.	NH_4TcO_4	$1.54 \pm 0.01_5$	0.1605±0.0003	0.1573±0.0006	3.69 ±0.01	5.80 ±0.03	3.77±0.04
	3	F.P.	NH_4TcO_4	0.603±0.006	0.0621±0.0004	0.0609±0.0004	3.69 ±0.01	2.25 ±0.02	3.72±0.05
	6	F.P.	Tc metal	0.909±0.005	0.0948±0.0003	0.0929±0.0004	3.69 ±0.01	$3.42_5 \pm 0.01_9$	3.77±0.03
	7	F.P.	Tc metal	0.909±0.005	0.0952±0.0003	0.0933±0.0004	3.69 ±0.01	$3.44_0 \pm 0.01_9$	3.78±0.03
	14	Mo	Tc metal[b]	0.559±0.008	0.0590±0.0001	0.0578±0.0002	3.69 ±0.01	$2.13_3 \pm 0.01_0$	3.82±0.06
	15	Mo	Tc metal[b]	0.559±0.008	0.0591±0.0002	0.0579±0.0003	3.69 ±0.01	$2.13_6 \pm 0.01_2$	3.82±0.06
	16	Mo	Tc metal[b]	0.559±0.008	0.0590±0.0002	0.0578±0.0003	3.69 ±0.01	$2.13_0 \pm 0.01_2$	3.81±0.06
Low absorption GM counter	1	F.P.	NH_4TcO_4	$1.54 \pm 0.01_5$	1.076 ±0.009		0.545±0.007	5.86 ±0.09	3.80±0.07
	3	F.P.	NH_4TcO_4	0.603±0.006	0.858 ±0.010		0.269±0.001	2.31 ±0.03	3.83±0.06
	16	Mo	Tc metal[b]	0.559±0.008	0.813 ±0.010		0.267±0.001	$2.16_8 \pm 0.02_8$	3.88±0.07
								Weighted mean	3.78±0.02

[a] F.P. =Tc from fission products; Mo =Tc from neutron irradiated molybdenum.

[b] Specific activity values low when based on metal weight. Tc weight values actually determined by absorption spectra analysis based upon standardization with F.P. material.

ratio at zero absorber (corrected for air and window) relative to the measured ratio with no absorber added.

It was also possible to use the absorption curves taken with the low absorption counters to determine the change in Tc/Co counting rate ratio with the addition of 1.13 mg/cm² absorber. Averaging the Fig. 3 extrapolation with the low absorption counter results, the correction factor was found to be 0.980±0.003. The data for the $Tc^{99}-Co^{60}$ comparisons are given in Table II. The Co^{60} disintegration rates were corrected for decay using 5.3 years as the half-life. The average value of the specific activity of Tc was found to be 3.78×10^4 dis/min-μg, which corresponds to a half-life of 2.12×10^5 years. The error is estimated to be ±2 percent.

V. COMPARISON WITH OTHER HALF-LIFE MEASUREMENTS

The earliest half-life values of Tc^{99} were based upon beta-activity measurements of samples extracted from neutron-irradiated uranium. From estimates of the chemical yield of extraction and assuming that the fission yield was about 6 percent, Lincoln and Sullivan[2] calculated a half-life value of about 10^6 years, while Schuman[3] quoted a value of 3×10^5 years (within a factor of five).

Two measurements on weighed samples were later made at Oak Ridge. Parker, Reed, and Ruch[25] isolated milligram amounts of technetium from fission products and precipitated and weighed technetium sulfide, assuming its formula to be Tc_2S_7 (by analogy to rhenium). Counting was carried out in a low geometry, low absorption counter. The aluminum absorption curve was extrapolated to zero absorber, neglecting the observed upswing at low absorber values. Their half-life value was $5.5 \pm 0.4 \times 10^5$ years.

Motta, Boyd, and Larson[5] isolated weighable amounts of technetium from neutron irradiated molybdenum. One hundred micrograms were electroplated on a copper disk, with the assumption that the deposit was technetium metal. The sample was counted with the usual mica window GM tube, comparing them with Co^{60} standards electroplated in a similar way. Their value was 9.4×10^5 years.

The specific activity determinations of Motta, Boyd, and Larson and Parker, Reed, and Ruch depend on an assumed composition for a particular technetium sample with no opportunity for cross checks. Parker, Reed, and Ruch, using technetium sulfide, were dealing with a particularly difficult compound, since it has been our experience that such sulfides carry varying quantities of free sulfur when precipitated from solutions.

Technetium, electrodeposited from solution, was assumed to be the metal and was weighed as such. Again there was no opportunity for cross checking and it would seem possible that inert material was carried down.

If the assumption is made that previous determinations were made on technetium containing inert impurities, it then becomes possible to reconcile the various values and with our value of 2.12×10^5 years. In view of the difficulties experienced in our work it seems to be a likely possibility.

VI. BETA-DECAY ENERGY OF Tc^{99} (FROM ABSORPTION CURVE)

In the course of investigating the radiochemical purity of the technetium sample, a careful absorption curve was taken (Fig. 1). The methane counter described in Sec. III was used, with aluminum absorbers placed against the window. Feather analysis[26] relative to RaE gave a range of 81.6 mg/cm². According to the

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[25] Parker, Reed, and Ruch, Atomic Energy Commission Document AECD-2043, January 9, 1948; Oak Ridge National Laboratory Report MonN-311, June 24, 1947 (unpublished).

[26] N. Feather, Proc. Camb. Phil. Soc. 34, 599 (1938).

revised Feather relationship[27]

$$E=1.92R^{0.725}\quad\begin{cases}E\ \text{in Mev}\\ R<0.3\ \text{g/cm}^2\ \text{Al.}\end{cases}\tag{4}$$

From this equation the energy of Tc^{99} is found to be 0.312 Mev, which agrees well with spectrographic values of 0.309 Mev[28] and 0.30±0.1 Mev.[29]

Previous results based on measurements of the initial half-thickness of the absorption curve, or on Feather analyses were 0.3 Mev,[2,3] 0.32 Mev,[4,5] and 0.31±0.03 Mev.[25]

The absorption measurements showed that the ratio of beta- to gamma-counting rates was greater than 20,000, which corroborates the previous measurements[3–5] indicating no gamma-ray. These results indicate less than 1 gamma-ray per 100 beta-particles.

We wish to express our gratitude to Mr. Laurence Sjoblom, Mrs. Marilyn Sietsema, Mrs. R. Hospelhorn, and particularly Mr. Jerome Lerner, who aided us at various stages of the measurements.

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[27] L. E. Glendenin and C. D. Coryell, "The relationship between the range and spectrograph energy of beta particles," paper 11, *Radiochemical Studies; The Fission Products* (McGraw-Hill Book Company, Inc., New York, to be published), National Nuclear Energy Series, Division IV, Vol. 9B; Nucleonics 2, 12 (January, 1948).

[28] M. Freedman and F. Wagner (to be published).

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The Reaction Energies of Light Nuclei from Magnetic Analysis*

E. N. STRAIT,† D. M. VAN PATTER, W. W. BUECHNER, AND A. SPERDUTO
Physics Department and Laboratory for Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts

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An extensive survey has been made of the reaction energies of ground-state transitions induced by proton and deuteron bombardment of light nuclei. A total of nineteen stable nuclei from deuterium through sulfur was investigated, and the Q-values directly measured for thirty-eight different reactions. From these Q-values, the energy releases of a number of other reactions have been computed. The reactions studied include the (p,α), (d,p), and (d,α); and the individual Q-values have been determined to precisions that are between 0.08 and 0.25 percent. The reaction products were analyzed by means of 180-degree focusing in the field of a large annular magnet.

The Q-values measured include those of a series of nuclear reactions which directly connect the mass of the alpha-particle with the deuteron mass and O^{16}. From these measurements, the mass of the alpha-particle has been determined as 4.003865±0.000007 amu.

I. INTRODUCTION

UNTIL the last few years measurements of the reaction energies of ground-state transitions involving charged particles were confined to range methods where the precision available was generally not better than 100 kev. In many cases the results of these measurements have been used to establish the masses of the nuclei involved in the reactions. However, magnetic analysis, as opposed to range measurements, makes possible an improvement in precision of approximately a factor of 10, with an attendant improvement in the accuracy to which masses can be established.

In 1947, a high resolution analyzing system was constructed at the M. I. T. High Voltage Laboratory for the specific purpose of determining Q-values of nuclear reactions to a high precision.[1] This consisted of a 90-degree deflecting magnet for analysis of the bombarding deuterons or protons and a large annular magnet for 180-degree focusing of the reaction products. During 1948 and 1949, a number of reaction energies were measured with greater accuracy than previously reported. The reactions investigated were as follows:[2–4] $Li^6(d,p)Li^7$ and $Li^7(d,p)Li^8$; $Be^9(d,\alpha)Li^7$ and $Be^9(d,p)Be^{10}$; and $C^{12}(d,p)C^{13}$ and $O^{16}(d,p)O^{17}$. During the past year, these investigations have been extended to a survey of over thirty-five reactions involving target nuclei from D^2 to S^{32}, including remeasurements of the reactions already reported. In some of these reactions, the ground-state group has not been previously observed. This paper will be confined to a report of the results on the ground-state transitions. The results on transitions to excited states of the residual nuclei will be described in other publications, some of which have already appeared in print.[5,6]

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* A report of these measurements was made at the Chicago Meeting of the American Physical Society, November, 1950. Phys. Rev. **81**, 315 (1951).

† Now at Northwestern University, Evanston, Illinois.

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[5] Buechner, Van Patter, Strait, and Sperduto, Phys. Rev. **79**, 262 (1950).

[6] Van Patter, Sperduto, Strait, and Buechner, Phys. Rev. **79**, 900 (1950).